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Miles , Richard and Dogariu, Arthur

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14. ABSTRACT
Research on the possibility of detecting magnetic anomalies remotely using laser excitation of a naturally occurring atomic or molecular species in air has focused on the use of Xe129. Although Xe129 is only present in the atmosphere at molar fractions of tens of parts per billion, it is the best candidate for magnetic detection since it has a nuclear spin of ½, and there is no electron spin to depolarize the nuclear spin. Molecules rapidly lose spin orienation through collisions, however Xe129, an inert atomic species, can remain spin polarized for up to seconds in air. This permits the measurement of the surrounding magentic field through the measurement of the spin nutation rate around the external field. The research has demonstrated that 10 part per million concentrations of Xe can be detected using 2+1 Radar REMPI, limited by background signals from oxygen. Higher detection sensitivity is anticipated through preparation of metastable Xe followed by 1+1 Radar REMPI. Spin polarization is to be achieved through multiphoton interactions with circular polarized laser light. Proper wavelengths for this multiphoton process have been generated through two photon pumped lasing in atmospheric pressure xenon, and excitation of selected Xe electronic states has been achieved.

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Magnetic Anomaly Detection by Remote Means

Richard B. Miles and Arthur Dogariu
Mechanical and Aerospace Engineering Department,
Princeton University, Princeton, NJ 08544
phone: (609) 258-5131 email: miles@princeton.edu

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ABSTRACT

Research on the possibility of detecting magnetic anomalies remotely using laser excitation of a naturally occurring atomic or molecular species in air has focused on the use of Xe^{129}. Although Xe^{129} is only present in the atmosphere at molar fractions of tens of parts per billion, it is the best candidate for magnetic detection since it has a nuclear spin of ½, and there is no electron spin to depolarize the nuclear spin. Molecules rapidly lose spin orienation through collisions, however Xe_{129}, an inert atomic species, can remain spin polarized for up to seconds in air. This permits the measurement of the surrounding magentic field through the measurement of the spin nutation rate around the external field. The research has demonstrated that 10 part per million concentrations of Xe can be detected using 2+1 Radar REMPI, limited by background signals from oxygen. Higher detection sensitivity is anticipated through preparation of metastable Xe followed by 1+1 Radar REMPI. Spin polarization is to be achieved through multiphoton interactions with circular polarized laser light. Proper wavelengths for this multiphoton process have been generated through two photon pumped lasing in atmospheric pressure xenon, and excitation of selected Xe electronic states has been achieved.

LONG-TERM GOALS

The long term goal for this project is to develop a high sensitivity remote magnetic sensing scheme that can function in the atmosphere near the ocean's surface. The approach is to explore the use of nonlinear optical methods for nuclear spin pumping, and high sensitivity radar detection methods for detecting spin state evolution. The research target is to identify a remote magnetic detection scheme that could measure magnetic fields in Earth's atmosphere at a standoff distance of at least one kilometer from the transmitter/receiver with a relative sensitivity of at least one nanotesla.

OBJECTIVE

The objective of this project is to determine if there are realistic methods that use atmospheric constituents to measure nano Tesla variations in the earth's magnetic field from a remote platform. Our approach is to explore the suggestion made by Prof. Happer[1], who pointed out that naturally occurring xenon 129 has a nuclear spin of ½ with a lifetime of seconds, and thus could potentially be a vehicle for magnetic field detection. The problem is that the concentration of xenon in the atmosphere is around 80 parts per billion and xenon 129 accounts for less than 1/3 of that. Thus the remote detection of xenon is a major impediment to the realization of this concept. Furthermore, the current method for producing hyper polarized xenon is based on spin exchange with rubidium, and that approach is not practical for stand off applications.

Our success with the development of Radar REMPI for the detection of parts per billion concentrations of nitric oxide in air[2] has provided an avenue toward the high sensitivity remote detection of xenon. Figure 1 shows the Radar REMPI remote detection concept. It combines laser selectivity with radar sensitivity. The laser is tuned to fall on a single or multiphoton resonance of the selected atom or molecule, by which selective ionization of that atom or molecule can be achieved. That ionization is then observed though radar scattering from the associated free electrons. In the case on nitric oxide, for example, a single photon at 226 nm overlaps a resonant state and a second photon at the same wavelength leads to ionization. This is termed 1+1 Radar REMPI. For xenon, two photons simultaneously excite a resonant two-photon transition and a third photon ionizes the atom, corresponding to 2+1 Radar REMPI.

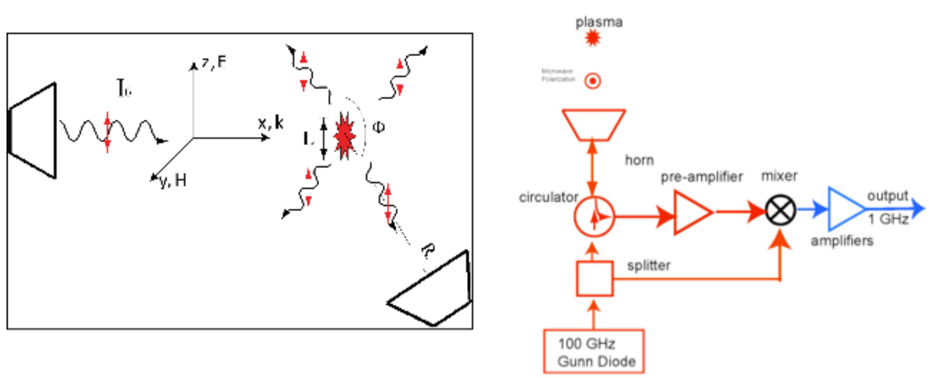

Figure 1: The Radar REMPI coherent homodyne detection process. Left – the laser created ionization region scatters microwave radiation. Right – the microwave scattering is routed to a pre amplifier and mixed with the original microwave source for coherent homodyne detection.

Figure 2 shows the relevant excited electronic states of xenon. The ground state is designated $1S_0$. The hyperfine separation of energy levels associated with electron coupling to the nuclear spin and the isotopic shifts are too small to be resolved at atmospheric pressures and temperatures. As a consequence, the selection of specific states must be achieved by controlling the polarization of the excitation laser(s).

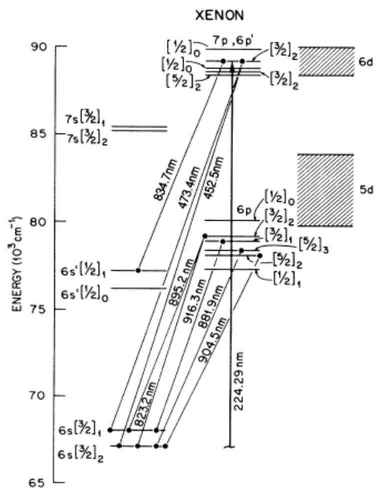

Figure 2: Xenon energy levels accessed by two photon absorption at 224-226 nm

Figure 3 shows the Radar REMPI spectra of four two-photon lines that fall between 224 and 226 nm, along with the designation of the upper level energy levels they are associated with. Note that the subscript refers to the J quantum number associated with the energy level. The J quantum number of the ground state is 0. Two photon excitation with linearly polarized light can satisfy $\Delta J = 0$, +/-1 and +/-2 selection rules, however two photon excitation with circularly polarized light can only satisfy the ΔJ = +/-2 selection rule, with the sign dependent on the sense of the polarization. Xenon 129 also has F quantum states, where **F=J+I**, and the F selection rules echo the J selection rules[3]. As a consequence the $7p[1/2]_0$ state is sensitive to the polarization, whereas the other three shown in Figure 3 are not. Figure 4 shows the reduction of the signal level of that state with circular polarization compared linear polarization, and contrasts that to the $7p[3/2]_2$ state, where there is no difference.

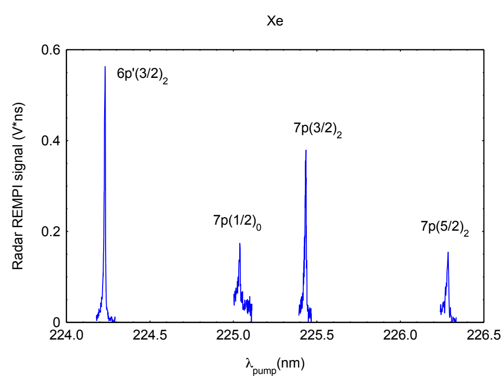

Figure 3: two photon resonant Radar REMPI lines of xenon.

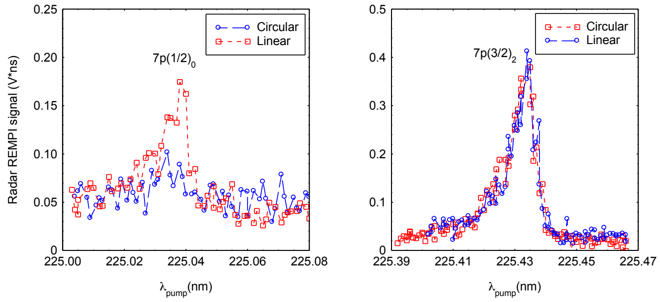

Figure 4: Circular vs. linear: $7p(1/2)_0$ is a polarization selective state, while $7p(3/2)_2$ is not

Measurements of the xenon concentration in air have been undertaken through step wise dilution, with the sensitivity achieved to date of ~10 parts per million. These data are shown in the left panel of Figure 5. The sensitivity is limited by the presence of two photon resonant molecular Rydberg states that overlap the xenon two photon resonance. The right panel shows that the measured signal is linearly related to the xenon density. Sensitivity can be increased through the use of double excitation pulses – one to selectively ionize the xenon and the second enhance the electron density associated with that ionization. Preliminary results show promise. Further increase in sensitivity is expected by preparing the xenon in the metastable state and then detecting it from that state with 1+1 Radar REMPI. This avoids the overlap with oxygen Rydberg states and provides spin selectivity through the three photon excitation of the metastable state. The detection of the precession of the xenon 129 is expected to be measurable though changes in the F projection (m state) following laser polarization.

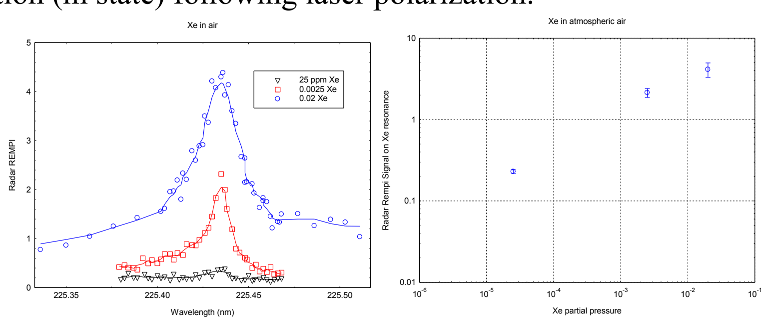

Figure 5: Detection of Xe in air via Radar REMPI. Left: Two-photon excitation spectrum to the $7p(3/2)_2$ state for traces of xenon in air. Right: Detection signal versus xenon density.

The process by which polarization is achieved must rely on optical pumping, which in this case involves resonant three photon pumping and four wave nonlinear processes. These lead to optical pumping and polarization of the ground state through two mechanisms: two photon resonant three photon pumping of the $6s'[1/2]_1$ state, and four wave generation of 129.5 nm radiation. An example of these processes is shown in figure 6. A 224.29 nm circularly polarized beam is two photon resonant with the ground state to $6p'[3/2]_2$ state transition and another co-propagating circularly polarized beam at 834.7 nm is resonant with the $6p'[3/2]_2$ state to $6s'[1/2]_1$ state transition (shown as solid red, green and blue lines in Fig. 6). These two co-propagating 224.29 nm and 834.7 nm beams drive atoms into the $6s'[1/2]_1$ state. This process is of interest because any atoms that begin in the ground m=+1/2 state are driven to the $6s'[1/2]_1$ m=+3/2 state from which they can only fluoresce to the m=+1/2 ground state (red circuit), so they are trapped in that state. Atoms in the m = -1/2 ground state, on the other hand, are driven to the m=+1/2 states of the 6s'$[1/2]_1$ state (blue and green circuits). From there they can fluoresce to the m=+1/2 or the m=-1/2 ground state. If they end up in the m=+1/2 state they are trapped. So through two photon resonant three photon optical pumping the m=+1/2 state is populated more than the m=-1/2 state, thus leading to polarization.

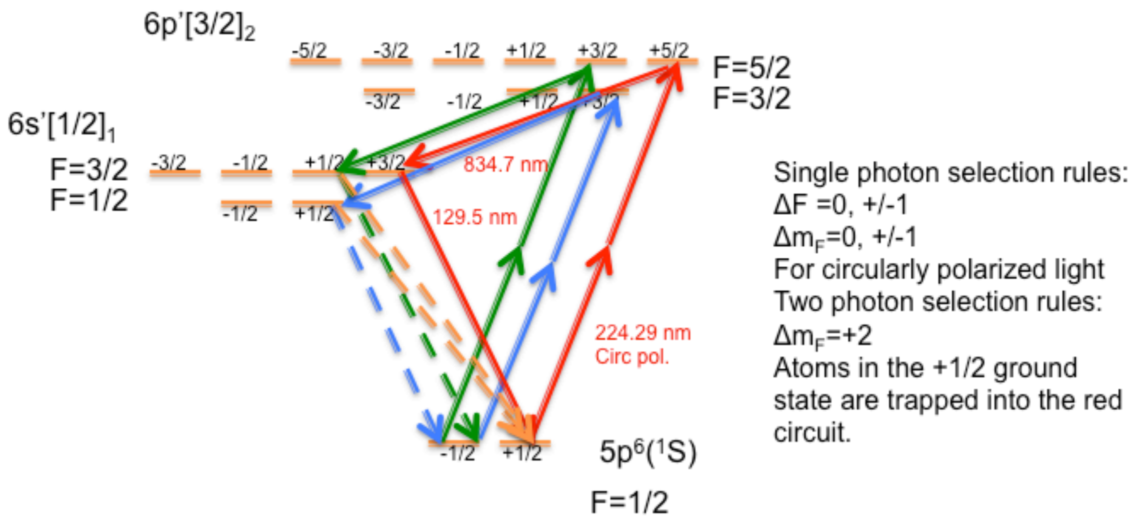

Figure 6: Possible method for double resonant four wave optical pumping of m=+1/2 ground state.

The nonlinear process also generates circularly polarized co propagating coherent light at 129.5 nm through resonant interactions with all xenon atoms and interactions with air molecules. The extinction length of this light at one atmosphere is about 3 mm. Atoms in the m=+1/2 state are pumped to the m=3/2 state, from which then can only relax back to the m=1/2 state. Atoms in the m=-1/2 ground state are pumped to the m=+1/2 excited state from which they can relax to either the m=1/2 or m=-1/2 state. Once they are in the m=+1/2 state, they are trapped leading to a more conventional single photon optical pumping by spontaneous radiation to the m=+1/2 ground state. This is shown in Figure 7 where the different colors represent different Δm transitions, but all are at 129.5 nm since the hyperfine spectra are overlapping. The dashed lines represent spontaneous emission and the solid lines represent the circularly polarized 129.5 driving radiation.

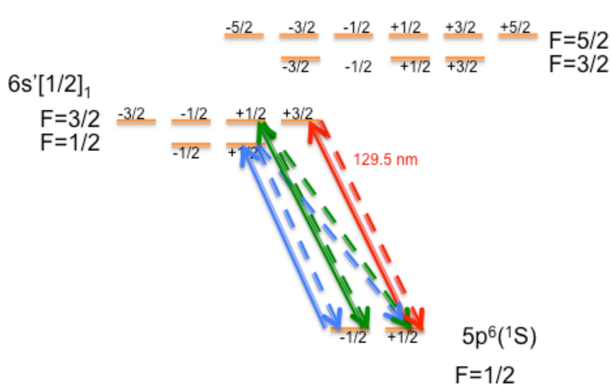

Figure 7: Single photon optical pumping of Xenon 129. The 129.5 nm coherent radiation is circular polarized.

WORK COMPLETED

We have successfully demonstrated spin selected two-photon resonant three photon pumping to achieve excitation of Xe atoms in an atmospheric pressure cell, and achieved two photon pumped lasing as a method to generate the third photon. We have shown that the polarization of that third photon can be controlled by the pump laser polarization. We have demonstrated the use of Radar REMPI to detect Xe using 2+1 Radar REMPI with various two photon resonances. We have also shown that double pulsing can reduce background noise through the dissociation of oxygen, and we have achieved detection of Xe down to 10 ppm in atmospheric air.

RESULTS

During this research effort we have demonstrated the detection of Xe using the remote Radar REMPI technique and the excitation of Xe atoms remotely using UV laser pulses. Specifically, we have achieved a detection concentration of Xe of about 10ppm in atmospheric air. One of the obstacles we are facing at very low concentration is the interference from non-resonant ionization of other constituents, in particular oxygen which has molecular two photon Rydberg state resonances that overlap the Xe transitions. We have learned that a double pulse pumping scheme can aleviate this problem by dissociating the molecular oxygen, and creating atomic oxygen instead, which shows no interference. We have also recognized that 1+1 Radar REMPI from an excited electonic state of xenon may significantly improved detection properties, and can be used in conjunction with spin selective Xe metastable state excitation.

Through tests with atmospheric pressure xenon, we have established a number of potential approahes for the spin polarizaton of Xe^{129} by resonant nonlinear three photon processes using circularly polarized laser beams that can be tranmittted through atmospheric pressure air. These processes can be made very efficient with short pulsed, high intensity lasers that are now available.

Two significant advances lead to very promising outcomes. The first is the three photon pumping of the Xe 6s'(1/2) J=1 state via two photon resonance with the 6p'(3/2) J=2 state using two photons at 224.31 nm and one at 834.7 nm. This process is followed by emission back to the ground state at 129.5 nm. The selection rules indicate that with circularly polarized light, this process will lead to spin polarizaton of the ground state of Xe^{129}. Our tests in atmospheric pressure xenon succeeded in demonstrating this pumping process, and, what is more interesting, the 834.7 nm beam is self generated inside the xenon, producing a well colimated laser output at 834.7 nm whose polarizaton reflects the selecton rules. This beam emerges synchronously and on top of the residual 224.31 nm pulse. Thus passing the driving two photon laser through an atmospheric pressure xenon cell automatically produces a beam with the correct three photons having the correct polarization, which, when focused into the air at a distance, will spin polarize the Xe^{129}.

The second advance is the three photon pumping of the 6s(3/2) J=2 metastable state via two photon resonant excitation of the 6p(3/2) J=2 state using two photons at 252.48 nm followed by a single photon at 823.5 nm. Here again the experiments have been done in atmospheric pressure pure xenon to establish the capability, and the 823.5 nm beam has been self generated within the atmspheric pressure cell, providing a well colimated output overlapping the residual 252.48 pump pulse in time and space. The polarization follows the pump polarization. The avantage of driving this metastable transition for detection is that the long lifetime of the metastable state may permit the subsequent 1+1 Radar REMPI detection to be relatively free from background interference from direct ionization of oxygen and other species.

IMPACT/APPLICATIONS

The impact of the work is that the potential for spin polarizing Xe^{129} in the atmosphere looks very promising. The challenge will be to detect the atoms with enough sensitivity to follow the nutation frequency.

RELATED PROJECTS

ONR supported research on "Backward Lasing in Air" (N00014-12-1-0090), that examined the potential for using various methods of backward lasing in air for the detection of trace species. Research has led to backward lasing using dissociation of molecular oxygen and molecular nitrogen followed by two photon exctation of the dissociated atomic oxygen and nitrogen, leading to backward lasing with photon efficiencies exceeding 0.1%. More recently ONR is supporting "Stand Off Detection of Trace Species by Simultaneous Radar REMPI and Backward Lasing" (N00014-15-1-2656).

REFERENCES

1. W. Happer, "Laser Remote Sensing of Magnetic Fields in the Atmosphere by Two-Photon Optical Pumping of Xe 129," , NADC Report N62269-78-M-6957 (1978)
2. A. Dogariu and R. B. Miles, "Detecting localized trace species using Radar REMPI," *Appl. Opt.* **50,** A68 (2011)
3. Keith D. Bonin and Thomas J. McIlrath, "Two Photon Electric Dipole Selection Rules," J. Opt. Soc. Am. B/Vol. 1, No. 1/March 1984

PUBLICATIONS

1. R. B. Miles and A. Dogariu, “"Flow Imaging and Standoff Detection by Dissociation of Air Molecules," in *Imaging and Applied Optics 2014*, OSA Technical Digest, LW2D.1 (2014).

2. A. Dogariu, T. L. Chng, and R. Miles, "Towards remote magnetic anomaly detection using Radar REMPI," in *CLEO: 2014*, OSA Technical Digest, SM4E.4 (2014).

Invited Presentations

1. Richard B. Miles and Arthur Dogariu , “Flow Imaging and Standoff Detection by Dissociation of Air Molecules,” Laser Applications to Chemical, Security and Environmental Analysis (LACSEA) 2014, Seattle, WA, July 13-17, 2014
2. Richard Miles, “Diagnostics by Dissociation; FLEET, lasing in air, and trace detection of complex molecules by Radar REMPI” AIAA Aviation 2014 Meeting, San Diego, CA, June15-18, 2014.
3. R. B. Miles, “Pumping Air: FLEET, Radar REMPI and Backward Lasing New Methods for Measuring Flow Properties and Contaminants in Air” Midwest Mechanics Seminar invited speaker
   Tour A: University of Michigan, Michigan State University, Notre Dame, Northwestern University, University of Wisconsin Oct. 12-16, 2015
   Tour B: Purdue, University of Illinois Urbana Champaign, Illinois Institute of Technology, Iowa State University, University of Minnesota Oct. 29-30, 2015

Conference Presentations

1. Tat Loon Chng, Richard B. Miles, “Understanding the Impact of Buffer Gases on the Radar REMPI Diagnostic” (AIAA 2015-2803) 46th AIAA Plasmadynamics and Lasers Conference, 2015, 10.2514/6.2015-2803
2. R. B. Miles “The Use of Radar for the Characterization of Laser-Generated Plasmas and for Stand-Off Trace Gas Detection”, 17th International Symposium on Laser-Aided Plasma Diagnostics Hokkaido, Japan, September 27 – October 1, 2015
3. R.B. Miles and A. Dogariu “Diagnostics by Dissociation: FLEET, lasing in air, and trace detection of complex molecules by Radar REMPI”, 2015 AIAA Aviation Forum, Atlanta GA. (June 17, 2014) (invited)
4. R. B. Miles, “Nonlinear Processes in Air,” International Conference on Coherent and Nonlinear Optics (ICONO 2013) International Conference on Lasers, Applications, and Technologies (LAT 2013) 2013, June 18-22, 2013 Moscow, Russia

STAFF SUPPORTED

Prof. Richard Miles, Robert Porter Patterson Professor Emeritus and Senior Scholar
Dr. Arthur Dogariu, Research Associate